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CORROSION PROPERTIES OF
RAPIDLY SOLIDIFIED COPPER BASE
ALLOYS FOR MARINE SERVICE

R.S. Hollingshead
S. Corbin - S.H. Lo

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R.S. Hollingshead - S. Corbin
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Approved by L.J. Leggat

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ABSTRACT

Copper base alloys are used in marine applications requiring a combination of high strength and corrosion resistance. In an effort to improve the performance of some selected copper base alloys, rapid solidification has been used to refine the microstructures and enhance solid solubilities. For this study, 90/10 CuNi, 70/30 chromium modified CuNi, 70/30 niobium modified CuNi and NiAl bronze alloys were rapidly solidified by meltspinning. A 90/10-8% Fe CuNi was similarly prepared. The corrosion behaviour of the rapidly solidified alloys was compared to conventionally processed ones with similar compositions, using electrochemical techniques.

It was found that the general corrosion resistance of the rapidly solidified alloys was as good as or better than the conventionally processed material, and that the rapidly solidified alloys may even be better under erosion/corrosion conditions. (Continued on page 1)

RESUME

Dans le domaine de la construction navale, les alliages de cuivre sont utilisés lorsqu'il faut à la fois une grande solidité et une excellente résistance à la corrosion. Dans le but d'améliorer les propriétés de certains alliages de cuivre, on a eu recours à la solidification rapide pour affiner les microstructures et pour augmenter la solubilité à l'état solide. On a rapidement solidifié, au cours du filage, des cupro-nickels (90/10) simples, alliés à du chrome (70/30), à du niobium (70/30), à du fer (90/10 - 8 Fe) ainsi que du bronze d'aluminium. La résistance à la corrosion des alliages ainsi traités a été comparée à celle d'alliages semblables à solidification classique par processus électrochimique. De manière générale, elle était équivalente ou supérieure; les alliages solidifiés rapidement pourraient offrir un rendement encore supérieur dans des conditions combinées d'érosion et de corrosion.



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INTRODUCTION

The metallurgist has long been aware of the improved properties of alloys that can be achieved by avoiding some structures predicted by equilibrium phase diagrams. The quenching of steels and the quenching and aging of aluminum alloys are both techniques which lead to dramatic improvements in properties. The advent of rapid solidification technology (RST) has enabled the metallurgist to avoid the undesirable structures predicted in equilibrium phase diagrams by an even greater degree and has resulted in some new alloy structures with even better mechanical and chemical properties.

Rapid solidification can be defined as the cooling of a metal or alloy from the molten to solidified state with cooling rates in the order of 10^5 to $10^7 \text{ K}^\circ/\text{sec}$. In the past, quenching had generally been applied to solid alloys with the result that cooling rates of about $3000 \text{ K}^\circ/\text{sec}$ were generated. It was soon evident, from preliminary studies of alloys produced by RST, that there were a number of structural features that could be produced.^{1,2} The most important of these are listed below:

1. Extension of solid state solubilities;
2. Refinement of the as cast grain size;
3. Modification of the segregation pattern;
4. Formation of metastable phases; and
5. Retention of alloy melt in a supercooled or glassy condition (formation of so called "metallic glass").

Alloys produced by RST can generally be split into two categories: (1) those alloys that are thermodynamically able to achieve a glassy condition upon being solidified with cooling rates in the range of 10^5 to $10^7 \text{ K}^\circ/\text{sec}$ and (2) those alloys that have microcrystalline structures.

The microcrystalline alloys exhibit a much finer and more homogeneous microstructure than conventional alloys. The smaller grain size will result in a higher yield strength for the alloy. For example, the Hall-Petch relation, $\sigma_o = \sigma_i + Ky D^{-1/2}$ where

σ_o = yield stress

σ_i = friction stress opposing motion of dislocations

K_y = measure of extent to which dislocations are piled up at barriers

D = grain diameter

shows that when the grain size reaches very small sizes, the effect on yield strength can be very substantial³. It should also be noted that heat treatment (i.e. precipitation hardening, etc.) can also improve the mechanical properties of these alloys. In addition to the mechanical properties, corrosion and oxidation resistance may also be enhanced, mainly due to the much finer and homogeneous structure.

RAPIDLY SOLIDIFIED COPPER BASE ALLOYS

The Defence Research Establishment Atlantic (DREA) has, for a number of years, carried out erosion corrosion studies on a number of copper base alloys for marine applications⁴. These include wrought 90/10 and 70/30 cupro nickels, cast NiAl bronze, and cast 90/10, 70/30 Nb modified and 70/30 Cr modified cupronickels. As part of a program to evaluate advanced materials and processing techniques, a project was initiated to compare the corrosion properties of "conventional" copper base alloys to similar compositions processed by rapid solidification techniques. Also, in order to take advantage of one of the benefits of RST, it was decided to study a 90/10 CuNi with 8% iron in the attempt to extend the solid solubility of iron (normally 2%) in CuNi to improve the alloy's corrosion resistance. This had also been attempted by Hammond⁵.

The majority of the work done in rapid solidification of both metallic glasses and microcrystalline alloys has been concentrated on superalloys and aluminum alloys. However, there have also been some studies on copper base alloys. Research on this alloy system has concentrated on binary systems of copper such as Cu-Al and Cu-Ni as well as Cu-Ni with additions of chromium, niobium and boron.

Yefimov, et al.⁶, studied the influence of cooling rates between 10^5 and 10^7 K/sec on a number of binary copper systems. They found that, in most cases, cooling rates of this magnitude could lead to suppression of peritectic reactions and monotectic phase separations, producing either a "pseudoeutectic" dispersed structure or a very fine-grained single-phased solid solution. Neither peritectic or monotectic alloys could be rendered amorphous at these cooling rates.

Yefimov, et al.⁶ also discovered that eutectic reactions could be suppressed in some cases, producing a single-phase, metastable solid solution of composition very close to the original molten alloy. When the cooling

rates exceeded 10^6 K⁰/sec and the original alloy had a composition very close to the eutectic point, an amorphous deposit could be obtained for certain alloys. They concluded that the cooling rates needed for suppression of the above reactions seemed to depend on a number of things including: (1) alloy system and location of elements on the periodic table, (2) the location of the alloy on the phase diagram, (3) the amount of melt overheating before quenching and (4) the impurity types and concentrations.

Panchanathan, et al.^{7,8}, studied Cu-Ni-B systems with varying amounts of aluminum and chromium additions. They found that, while conventional alloys containing these compositions show a high degree of segregation, similar alloys that are rapidly solidified and heat treated contain uniformly dispersed borides. This resulted in improved mechanical properties. For example, RST alloys attained a hardness of 500 Kg/mm², equivalent to a tensile strength of 1551 - 1723 MPa. In contrast, conventional copper-nickel alloys have tensile strengths of 310 - 620 MPa. In addition, although no experimental evidence was presented, Panchanathan stated that these alloys also exhibited good corrosion resistance.

Hammond⁵, on the other hand, evaluated the corrosion resistance of some meltspun CuNi alloys (90/10, 70/30 and 70/30 chromium modified) and found that the alloys readily corroded during electrochemical anodic polarization scans while immersed in aerated and deaerated solutions of 0.1M NaCl.

Other studies⁹ have shown that 90/10 and 70/30 copper nickel alloys with additions of nickel, iron, titanium and/or zirconium, produced by RST, exhibited greatly improved tensile properties over their conventional counterparts. In addition, a 3.2% chromium 70/30 copper nickel RST powder compact showed better corrosion resistance than its conventional counterpart.

Rapidly solidified aluminum bronzes were produced by Collins, et al.¹⁰. They found that improved strength and corrosion resistance over conventional alloys was evident in the RST material. These alloys exhibited a significantly refined microstructure and extended solid solubility of iron compared to the as cast bronze.

In summary, previous studies on the rapid solidification of copper base alloys reveal two things. First, upon rapid solidification, most of the alloys had some or all of the microstructural features described above. Second, all of the alloys showed improved mechanical strength and a large percentage of them also showed improved corrosion resistance in comparison to conventionally processed materials.

EXPERIMENTAL PROCEDURE

The rapidly solidified material used in this study was produced for DREA under contract by the Ontario Research Foundation, Mississauga, Ontario. The technique used to produce the RST material was melt spinning and is described by Lo, et al.¹¹. The mechanical properties are also described by these authors. The material was received by DREA in ribbon form (coated to prevent oxidation). Table I shows the compositions of the alloys used.

The RST ribbon was prepared by cutting it into lengths of about 2-3 cm² and placing these lengths into acetone to remove the coating. All sample preparation was performed within one hour prior to immersion in the electrolyte. The samples were then tested with no further preparation since the side that had touched the rotating cooling block had a mirror-like appearance. The conventional materials were also cut into the same sized sections but were further polished to 600 grit with SiC paper. Both the RST and parent materials were degreased in boiling toluene for 5 minutes.

The electrolyte used was natural seawater obtained from the Halifax Harbour. The seawater was filtered to 3 microns and its measured pH ranged between 7.5 and 7.9. The electrolyte was maintained at 25°C and was aerated for 20-30 minutes prior to testing and continued to be aerated throughout the test.

The sample holder used was a flat specimen holder which exposed a sample surface of 1 cm² to the electrolyte. The polarization resistance measurements were conducted in an EG and G Princeton Applied Research (PAR) Model K47 corrosion cell (Figure 1). The cell accommodates two high density counter electrodes, a working electrode (sample), a reference electrode bridge tube (luggins probe) with a fitted calomel reference electrode and a gas inlet, outlet purge tube.

The corrosion potential and corrosion current density of the copper base alloys were determined over a period of time. The experiments were conducted and the polarization resistance data were collected by a microprocessor controlled EG and G PAR Model 350 corrosion measurement system.

The measurements were taken 5 to 10 minutes after immersion of the sample in the electrolyte and approximately every two hours afterwards until termination of the test. The potentiodynamic scan was started 25mV cathodic to the sample's corrosion potential, which was measured by the instrument immediately before scanning and continued at a scan rate of 0.167 mV/sec. The

current required to maintain the sample at its controlled potential was measured every 0.6 mV throughout the linear scan. The 50 mV scan was completed 25 mV anodic to the sample's corrosion potential.

After a completed run, the current/potential data produced were stored on digital tape by an EG and G PAR Model 355 Tape Storage Module and then transferred through the 350 corrosion measurement system to a DREA DEC-20 computer through the use of the fortran program <STAAL> C11A20. The polarization resistance data were analyzed by the method of nonlinear least squares using a Pascal computer program called CORROS. The method of analysis used and both programs CORROS and <STAAL> C11A20 are described in detail by Hanham, et al.¹². Basically, CORROS is a program which calculates the corrosion current density (ICORR) by the polarization resistance method.

Galvanic corrosion studies were carried out using an EG and G PAR Model 351 corrosion measurement system. The study included two couples of 90/10 copper-nickel material: (1) parent/parent couple and (2) parent/RST-8% Fe couple. The experimental conditions used were the same as those used in the polarization resistance technique.

RESULTS AND DISCUSSION

The results of the corrosion rate measurements (Figures 2 - 5) indicate that the general corrosion resistances of the RST alloys are as good as or better than the conventionally cast materials. As well, the results showed some interesting differences in the responses of the two types of alloys immersed in sea water, that could result in the RST products being superior under erosion/corrosion conditions. The corrosion potential vs. time plots that were made showed that all of the RST alloys tended to passivate much quicker than the conventional ones (Figures 6 - 9). This can also be seen to a certain extent in the plots of corrosion rates vs. time. This would tend to indicate the formation of a much more stable passive film which could result in alloys being more resistant to erosion/corrosion. The reason for the more rapid passivation is probably related to both the much finer grain size and improved homogeneity. Figures 10-15 compare the grain sizes of cast and RST alloys. Whereas both the 70/30 alloys and the 90/10 are essentially solid solutions (the Cr modified alloy does have some coherent precipitation hardening¹³), the NiAl bronze has a complicated metallurgical structure which significantly affects the corrosion resistance of the alloy¹⁴.

The microstructure of the conventionally cast NiAl bronze alloy can be very complex. The various phases that can be present are the α matrix, the γ_2 phase and the κ phases. There can be various forms of the κ phase (κ_i , κ_{ii} , κ_{iii} , κ_{iv}). Dealuminification can occur because of a galvanic action (on a microscale) between the α and γ_2 , and the α and κ phases. The metallographic examination of the conventional NiAl bronze (Figure 16) shows the presence of α (light region), κ_{iii} (lamellar), an NiAlFe rich phase, and κ_{ii} (dark globular particles), an FeAl rich phase. Figure 17 shows the structure of the RST material with only the α phase apparently present. Consequently, the improvement of the corrosion resistance is due to the lack of γ_2 and κ phases and improved homogeneity.

It has been known for many years that the addition of iron to the 90/10 CuNi will improve its corrosion resistance. Popplewell, et al.¹⁵, found that when 90/10 CuNi was modified with iron, doping of Cu_2O corrosion product by iron ions significantly increased the electronic resistance of the film and also decreased the corrosion rate. Iron is normally only soluble to about 2% in copper: above 2% an iron rich phase begins to precipitate out. It has been found¹⁶ that precipitation of this phase discontinuously will reduce the alloy's corrosion resistance. Figure 2 shows that there was an improvement in the corrosion resistance of the RST alloy containing the 8% iron, and it appears that this alloy was the quickest to passivate (Figure 6) of those evaluated. When comparing microstructures (Figures 18 & 19), it can be seen that there was no apparent precipitation of the second phase. All of the iron is probably in solid solution with a commensurate increase of iron in the Cu_2O surface film. Hammond⁵ found that for similar compositions there was no difference in the corrosion rate between RST and conventional products. However, he indicated that he did not get all of the iron into solid solution and that there was a definite second phase. A confirmation of the improved corrosion resistance of RST material 90/10-8% Fe was found in the measurements of galvanic action. Figure 20 shows that there is a potential difference between the RST and conventional or parent alloy where the RST alloy became the cathode and the conventional alloy became the anode. This potential difference resulted in a galvanic current (Figure 21).

Further corrosion studies of the RST copper alloys are presently underway, but they involve material made from RST powder produced by water

atomization and compacted by hot extrusion and hot isostatic pressing¹⁷. The studies will include corrosion measurements with a rotating electrode to simulate flowing conditions (erosion/corrosion) in a sea water system.

CONCLUSIONS

The results of this study demonstrate that rapid solidification of selected copper base alloys results in products which have a general corrosion resistance in an aerated sea water environment which is as good as or better than that of conventionally processed alloys. All of the RST alloys tended to passivate much quicker than the conventional alloys, with what is suspected to be a much more stable tenacious passive film which would likely make these alloys more resistant to erosion/corrosion.

The study also showed it is possible by RST to eliminate undesirable phases in NiAl bronze and improve its corrosion resistance. Finally, it was shown that it was possible by RST to extend the range of the solid solubility of iron in 90/10 CuNi alloys and improve this alloy's overall general corrosion resistance.

TABLE 1
CHEMICAL COMPOSITION OF COPPER NICKEL ALLOYS

	<u>Cu</u>	<u>Ni</u>	<u>Fe</u>	<u>Mn</u>	<u>Cr</u>	<u>Nb</u>	<u>A1</u>
90/10	BAL	10.5	1.42	0.41			
90/10 - 8 Fe	BAL	12.1	8.50	0.003			
70/30 Cr	BAL	30.0	0.75	0.63	1.7		
70/30 Nb	BAL	28.9	0.77	0.52		0.9	
NiAl Bronze	BAL	5.7	4.50	0.58			9.2

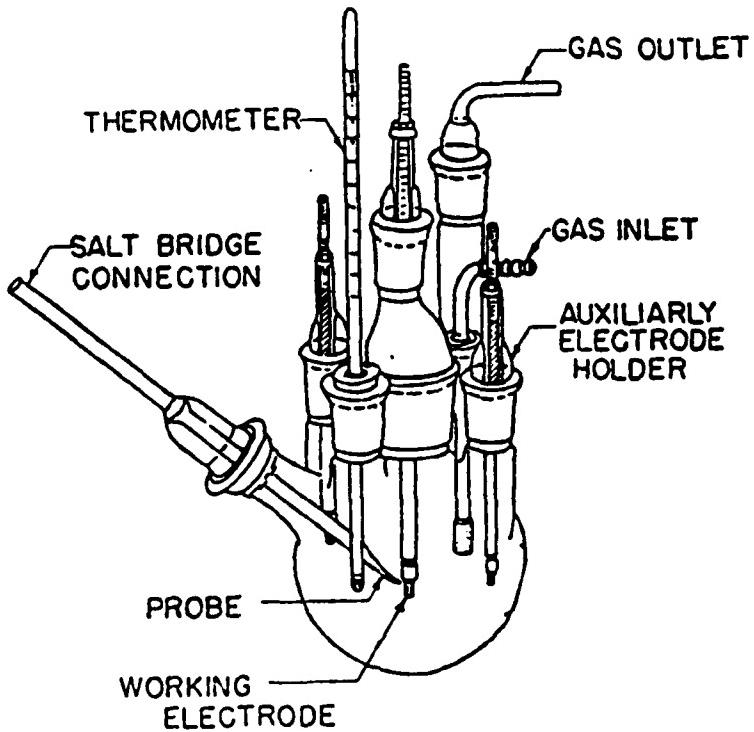


Figure 1. Standard Polarization Cell Schematic Diagram.

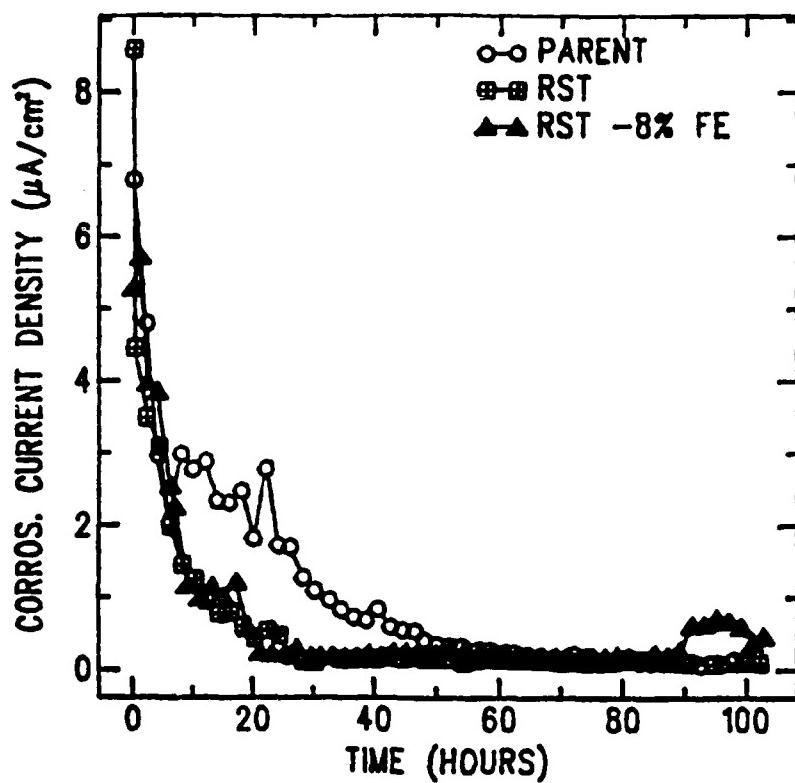


Figure 2. Corrosion Rate vs Time For 90/10 Alloys in Aerated Seawater.

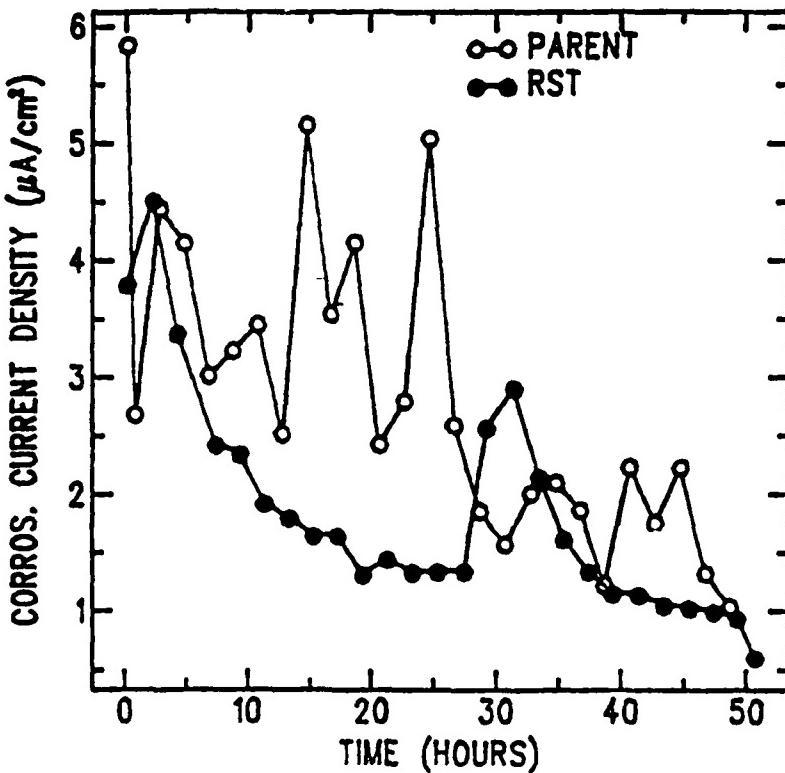


Figure 3. Corrosion Rate vs Time For 70/30 Nb Mod. Alloys in Aerated Seawater.

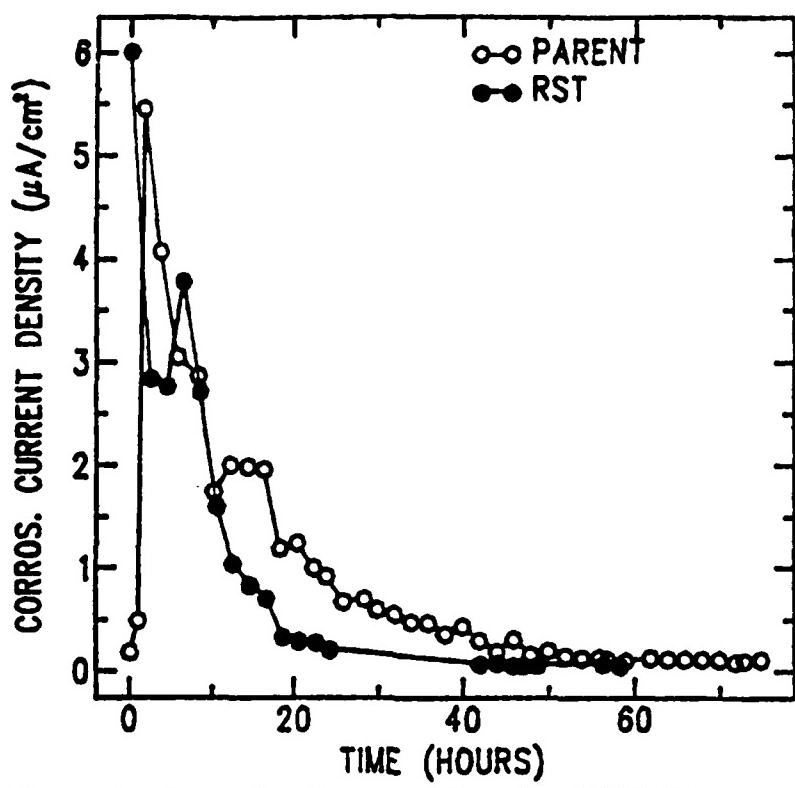


Figure 4. Corrosion Rate vs Time For 70/30 Cr Mod. Alloys in Aerated Seawater.

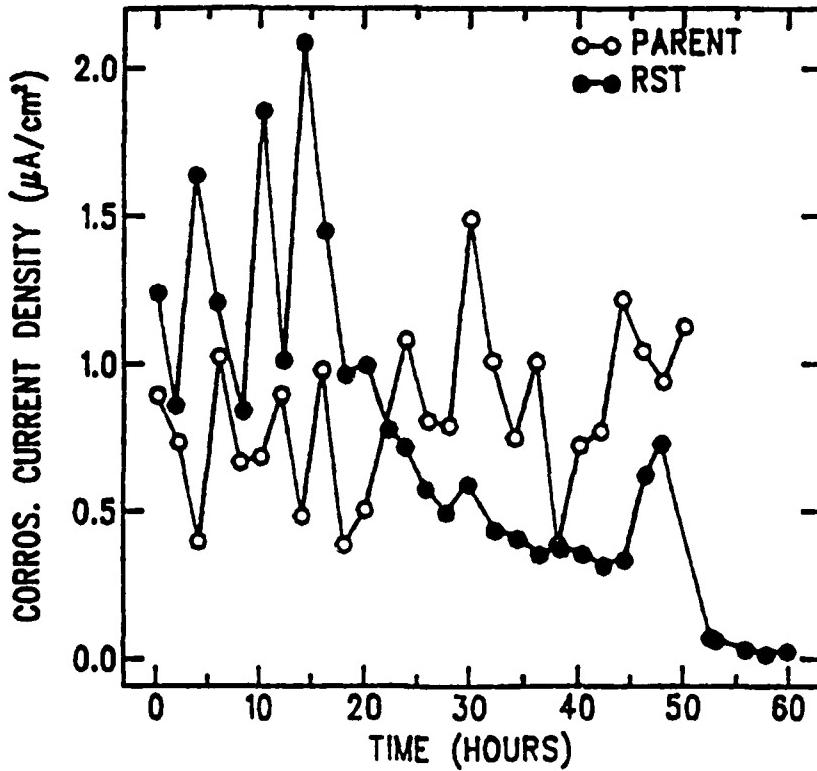


Figure 5. Corrosion Rate vs Time For NiAl Bronze Alloys in Aerated Seawater.

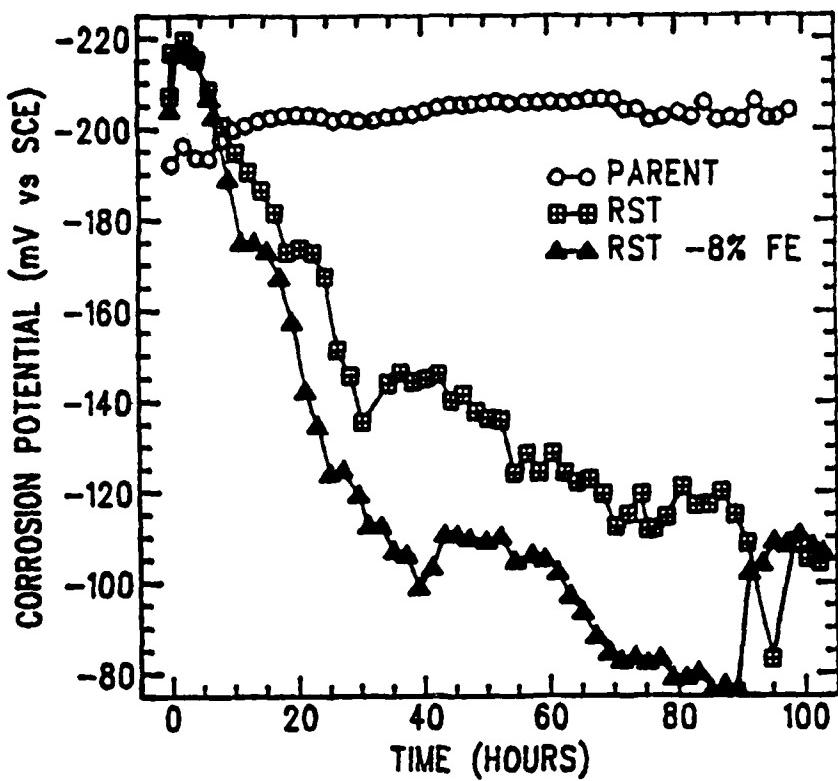


Figure 6. Corrosion Potential vs Time For 90/10 Alloys in Aerated Seawater.

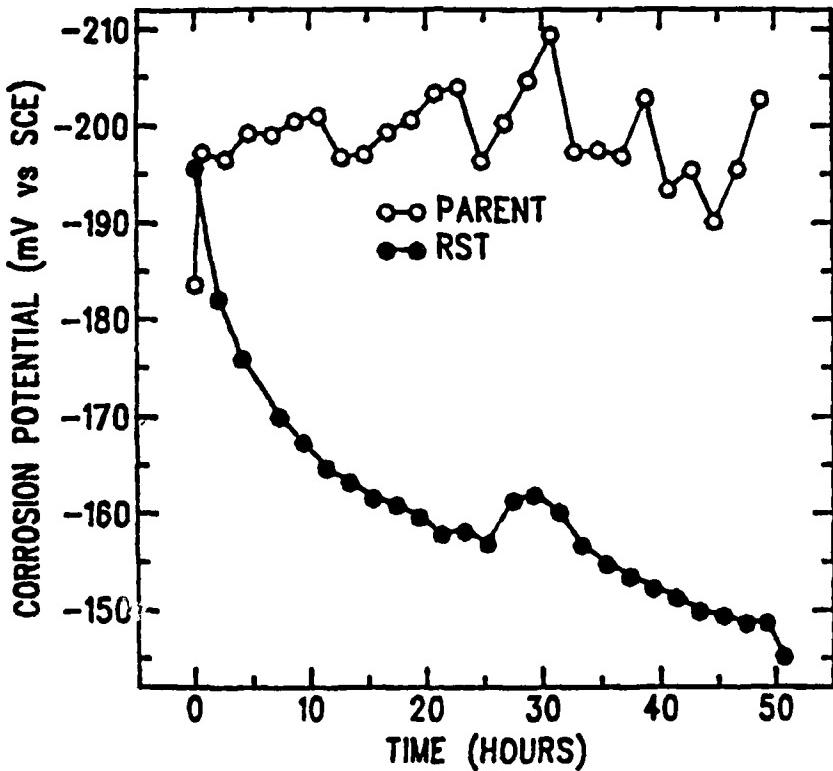


Figure 7. Corrosion Potential vs Time For 70/30 Nb Mod. Alloys in Aerated Seawater.

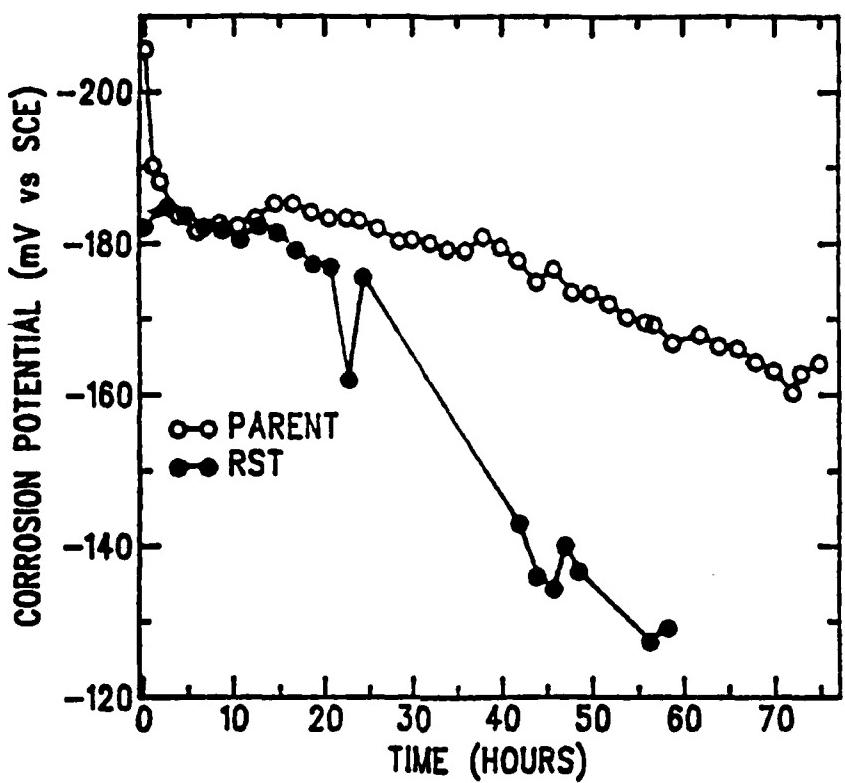


Figure 8. Corrosion Potential vs Time For 70/30 Cr Mod. Alloys in Aerated Seawater.

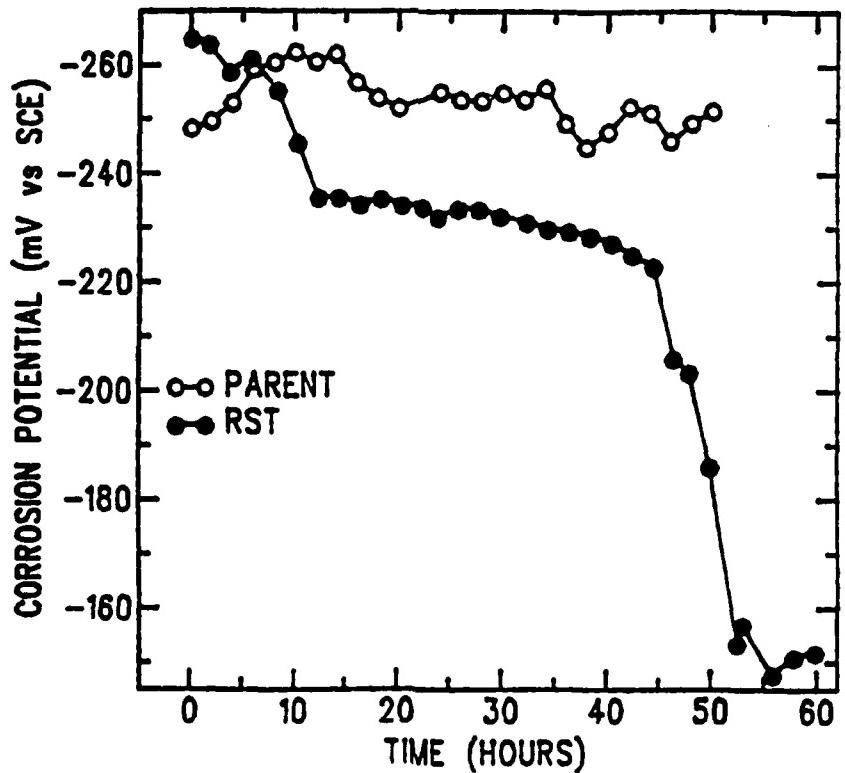


Figure 9. Corrosion Potential vs Time For NiAl Bronze Alloys in Aerated Seawater.



FIGURE 10: As cast microstructure of 90/10 alloy showing large dendritic grain size.¹⁸ 50X.

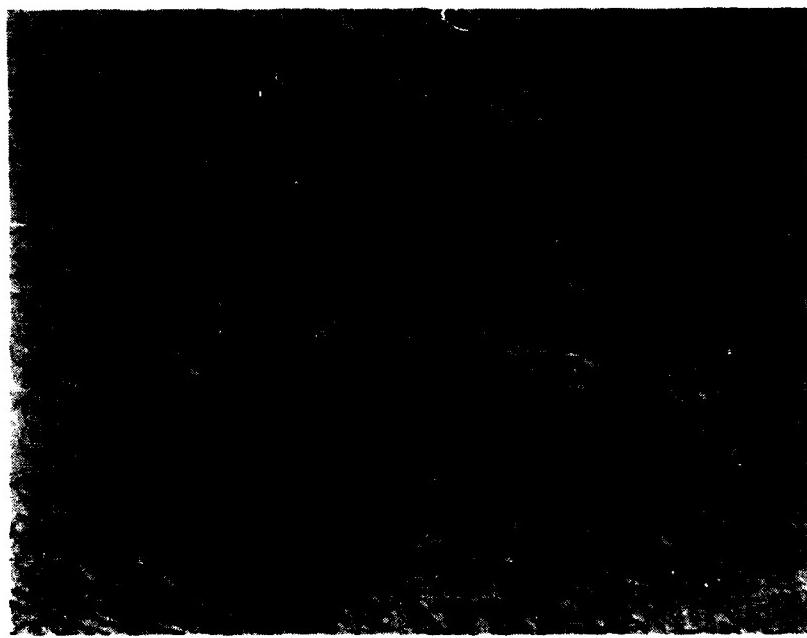


FIGURE 11: RST microstructure 90/10 alloy showing fine grain size.¹⁸ 1000X.

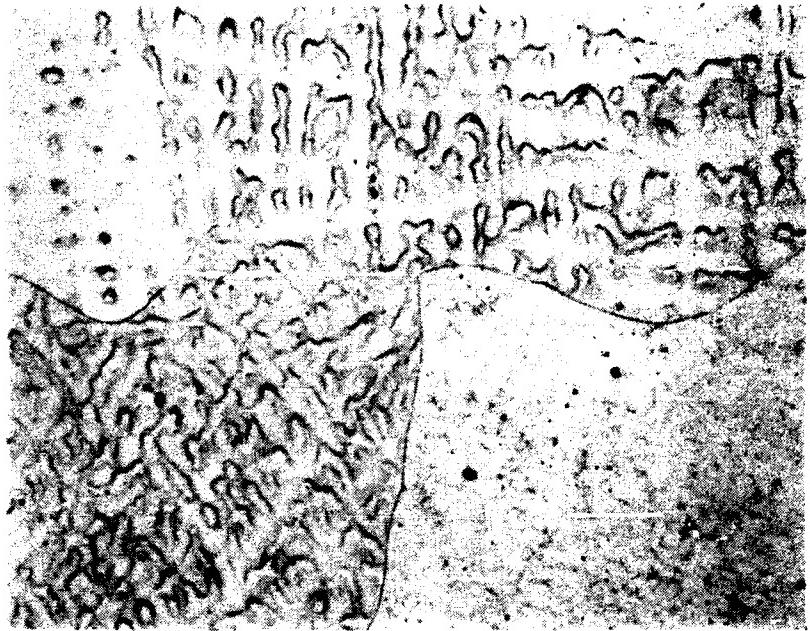


FIGURE 12: As cast microstructure of 70/30 Nb modified alloy showing large dendritic grain size.¹⁸ 200X.



FIGURE 13: RST microstructure 70/30 Nb modified alloy showing fine grain size.¹⁸ 1000X.



FIGURE 14: As cast microstructure of 70/30 Cr modified alloy showing large dendritic grain size.¹⁸ 200X.



FIGURE 15: RST microstructure 70/30 Cr modified alloy showing a fine grain size.¹⁸ 1000X.



FIGURE 16: As cast microstructure of NiAl bronze showing α phase (white), κ_{iii} (lamellar) and κ_{ii} (globular). 900X.

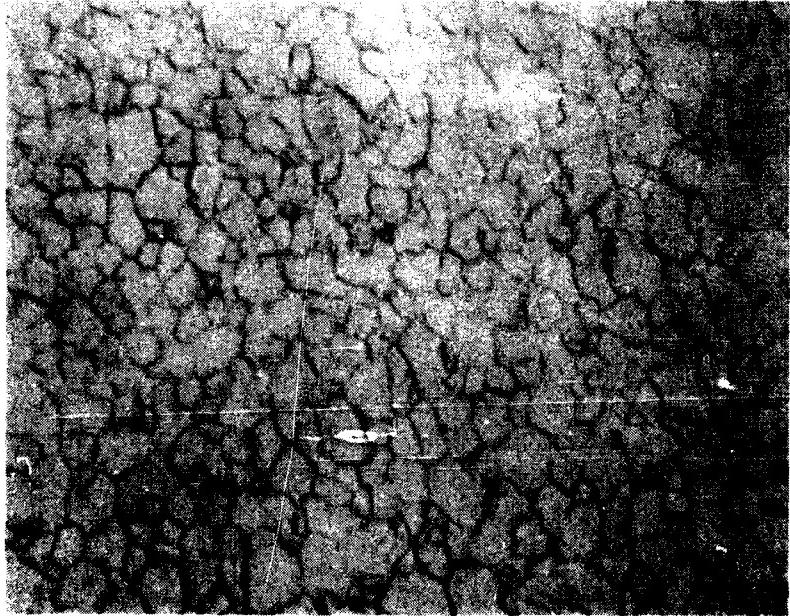


FIGURE 17: RST microstructure of NiAl bronze showing α phase with no apparent second phases.¹⁸ 1600X.



FIGURE 18: Microstructure of cast 90/10 - 8% Fe showing large grain size and an Fe rich second phase.¹⁸ 500X.



FIGURE 19: RST microstructure of 90/10 - 8% Fe showing fine grain size and no apparent second phase.¹⁸ 1000X.

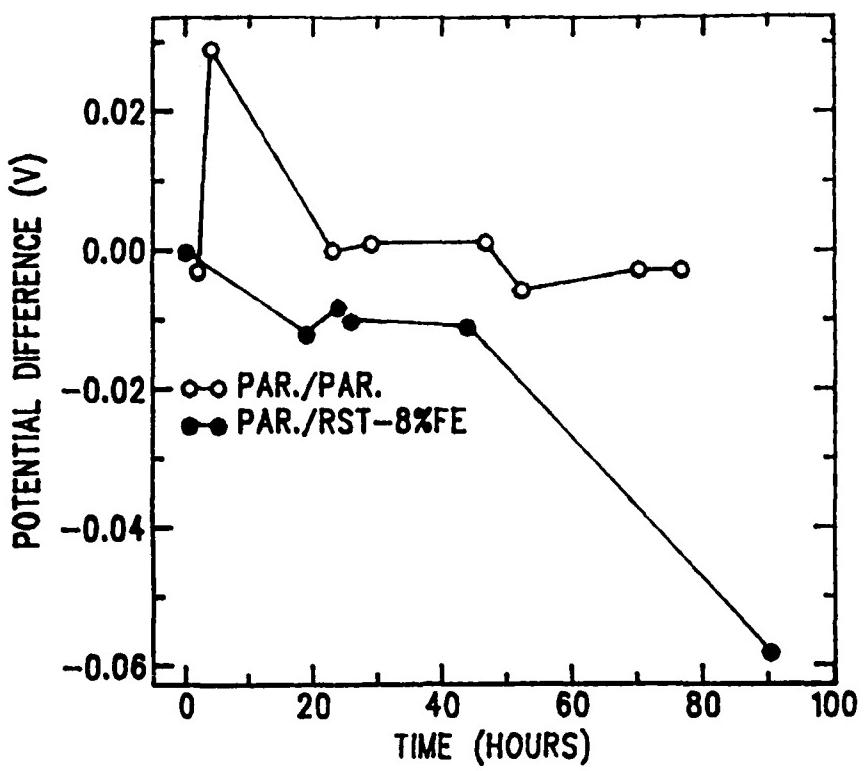


Figure 20. Galvanic Potential Differences of 90/10 Couples in Aerated Seawater.

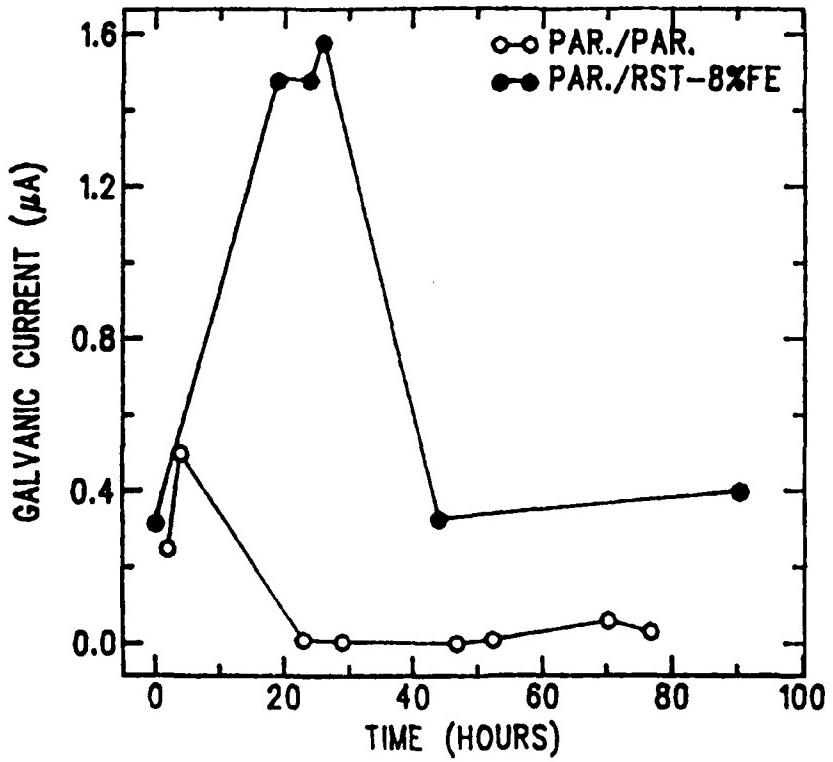


Figure 21. Galvanic Current of 90/10 Couples in Aerated Seawater.

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Copper base alloys are used in marine applications requiring a combination of high strength and corrosion resistance. In an effort to improve the performance of some selected copper base alloys, rapid solidification has been used to refine the microstructures and enhance solid solubilities. For this study, 90/10 CuNi, 70/30 chromium modified CuNi, 70/30 niobium modified CuNi and NiAl bronze alloys were rapidly solidified by meltspinning. A 90/10-8% Fe CuNi was similarly prepared. The corrosion behaviour of the rapidly solidified alloys was compared to conventionally processed ones with similar compositions, using electrochemical techniques.

It was found that the general corrosion resistance of the rapidly solidified alloys was as good as or better than the conventionally processed material, and that the rapidly solidified alloys may even be better under erosion/corrosion conditions.

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